

# On the evaluation of the ac-Stark shift and Kramers-Heisenberg dispersion formula

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We describe a unified approach for the determination of ac-Stark correction and Kramers-Heisenberg dispersion formula. In both cases the contribution from infinite intermediate summation appearing in the expression for the corresponding matrix elements are evaluated exactly in the dipole approximation for the ground state of hydrogen atom using a variation of the Dalgarno-Lewis method. The analytical expressions obtained can be efficiently used for the numerical evaluation of matrix element for all values of incident photon energy and comparison is made with results obtained by different methods.

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## I. INTRODUCTION

In the presence of intense laser field, atomic or molecular levels are displaced (shifted or broadened) and these stimulated radiative corrections are alternatively known as ac-Stark shift, dynamic Stark effect or light shift [1]. Being a fundamental process ac-stark effect is very well studied theoretically as well as experimentally in wide area of atomic and molecular physics. This effect is also very important in the area of laser trapping and cooling and is a basic mechanism behind many nonlinear optical effects [2]. It is a promising tool for the field of optical communications (Optical switching methods) and plays a very decisive role in many current research areas [3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14]. In this work we have described an efficient method to obtain a closed form analytical expression for two of the basic process viz. ac stark shift and elastic scattering cross section of photons for the atomic hydrogen in the ground state. Our aim is to get an analytical expression for dipole dynamical polarizability  $\tau^{(2)}(\omega)$  from which one can calculate the level shift  $\delta E^{(2)}$  and level width  $\Gamma^{(2)}$ . The hydrogen atom being the simplest of all atomic systems, plays a very special role in this respect because it has closed-form wavefunctions. Since these wave functions are analytically known, both the dipole dynamic polarizability and Kramer's-Heisenberg dispersion formula for hydrogen atom can be written in a closed form.

The level shift for atomic hydrogen was previously calculated by both perturbative and nonperturbative methods [15, 16, 17, 18, 19, 20, 21, 22, 23, 24]. In the perturbative method, evaluation of the higher order matrix element is the key problem and it is achieved through Greens function formalism [16, 17, 18] or by solving a system of second-order differential equations [20].

The Kramers-Heisenberg formula alternatively known

as dispersion formula plays a very important role in the determination of scattering cross-sections [25]. Considering its importance as an age-old relation there were only very few attempts to evaluate it analytically [26, 27]. There were some generalization of it and alternate forms are also available in the literature [29, 30, 31, 32]. Recently a low-energy expansion of the Kramers-Heisenberg formula for Rayleigh scattering process in atomic hydrogen was reported [33]. Similar to the ac Stark shift this also involves the evaluation of the second order matrix element.

The exact calculation of higher order process in the perturbative formulation is nontrivial because of the presence of infinite summation over intermediate states in the expression for the higher order matrix element. In our approach the matrix element containing infinite summation over the whole hydrogenic spectrum (discrete and continuum) is performed by using an implicit summation technique due to Dalgarno Lewis [34], which reduces the evaluation of the infinite summation to finding a solution of some inhomogeneous differential equation. The closed form expression which we have obtained is very simple and also very convenient for analytical continuation. Thus with this method, we can very easily obtain the relevant matrix element for radiation with energy larger than ionization energy (above threshold ionization), while other methods need some kind of approximation like Pade approximation in the case of Coulomb Greens function formalism.

## II. RADIATIVE CORRECTION IN THE GROUND STATE

The level shift depends on the intensity  $I$  and the frequency  $\omega$  of the radiation and the complex second order shift  $\Delta^{(2)}$  is given by [17, 18, 19, 20]

$$\Delta^{(2)}(\omega) = \delta E^{(2)} - i\Gamma^{(2)} = -\frac{I}{I_0}\tau^{(2)}(\omega) \quad (1)$$

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where the real part  $\delta E^{(2)}$  is the energy shift and the imaginary part  $\Gamma^{(2)}$  gives the level width and  $I_0 = 7.016 \times 10^{16}$  W/cm<sup>2</sup> is the characteristic atomic field strength intensity. Here  $\tau^{(2)}(\omega)$  is the dipole dynamic polarizability and in the atomic unit it can be written as

$$\tau^{(2)} = \sum_n \left\{ \frac{\langle g | \epsilon^* \cdot \mathbf{r} | n \rangle \langle n | \epsilon \cdot \mathbf{r} | g \rangle}{(E_g - E_n + \omega)} + \frac{\langle g | \epsilon \cdot \mathbf{r} | n \rangle \langle n | \epsilon^* \cdot \mathbf{r} | g \rangle}{(E_g - E_n - \omega)} \right\} \quad (2)$$

where  $E_g$  is the atomic ground state energy and  $\epsilon$  is the polarization of the radiation. First term in the bracket is the absorption-emission term and the second is the emission-absorption term. The infinite summation over the complete set of intermediate state in equation (2) can be performed exactly by defining a set of operators  $F$  and  $\tilde{F}$  such that

$$\epsilon \cdot \mathbf{r} |g\rangle = (FH_0 - H_0F + \omega F) |g\rangle \quad (3)$$

$$\epsilon^* \cdot \mathbf{r} |g\rangle = (\tilde{F}H_0 - H_0\tilde{F} - \omega\tilde{F}) |g\rangle \quad (4)$$

where  $H_0 = -\nabla^2/2 - 1/r$  is the unperturbed Hamiltonian for atomic hydrogen and  $|g\rangle = e^{-r}/\sqrt{\pi}$  is the ground state wave function in atomic units. With these definitions and the closure relation  $\sum_n |n\rangle \langle n| = \hat{I}$  the expression in equation (2) for dipole dynamical polarizability will be reduced to

$$\tau^{(2)}(\omega) = \langle g | \epsilon^* \cdot \mathbf{r} F | g \rangle + \langle g | \epsilon \cdot \mathbf{r} \tilde{F} | g \rangle. \quad (5)$$

Thus the infinite summation over the intermediate states are reduced to the determination of the operator  $F$  and  $\tilde{F}$ . Using co-ordinate space representation and writing  $F = \epsilon \cdot \mathbf{r} f(r)$  and  $\tilde{F} = \epsilon^* \cdot \mathbf{r} \tilde{f}(r)$ , the equations (3) and (4) become

$$r \frac{d^2}{dr^2} f(r) + (4 - 2r) \frac{d}{dr} f(r) + (2\omega r - 2)f(r) = 2r \quad (6)$$

$$r \frac{d^2}{dr^2} \tilde{f}(r) + (4 - 2r) \frac{d}{dr} \tilde{f}(r) - (2\omega r + 2)\tilde{f}(r) = 2r \quad (7)$$

By the method of Laplace transform [36] for the solution of differential equation we can obtain the solutions to the above differential equations as

$$f(r) = \frac{1}{\omega} - \frac{1}{2\omega^3} \Phi(1, 1, \lambda, r) \quad (8)$$

and

$$\tilde{f}(r) = \frac{1}{\omega} + \frac{1}{2\omega^3} \tilde{\Phi}(1, 1, \tilde{\lambda}, r) \quad (9)$$

with

$$\Phi(p, q, \lambda, r) = \int_{\lambda}^1 ds e^{-r(s-1)} K(p, q, \lambda, s) \quad (10)$$

$$\tilde{\Phi}(p, q, \lambda, r) = \int_1^{\lambda} ds e^{-r(s-1)} \tilde{K}(p, q, \lambda, s) \quad (11)$$

$$K(p, q, \lambda, s) = \left( \frac{1-\lambda}{1+\lambda} \right)^{\frac{1}{\lambda}} (s+\lambda)^{p+\frac{1}{\lambda}} (s-\lambda)^{q-\frac{1}{\lambda}} \quad (12)$$

$$\tilde{K}(p, q, \lambda, s) = \left( \frac{\lambda-1}{\lambda+1} \right)^{\frac{1}{\lambda}} (\lambda+s)^{p+\frac{1}{\lambda}} (\lambda-s)^{q-\frac{1}{\lambda}} \quad (13)$$

where  $\lambda = \sqrt{1-2\omega}$  and  $\tilde{\lambda} = \sqrt{1+2\omega}$ . In this work we obtain the solutions with integer values of  $p$  and  $q$ . But in general for the purpose of analytic continuation it can be complex and  $\lambda$  also can become complex depending on the value of the frequency  $\omega$ . Now using equations (10) and (11) in equation (5) the final form of the dipolar polarizability becomes

$$\tau^{(2)}(\omega) = \frac{2}{3\omega^3} \int_0^{\infty} dr e^{-2r} r^4 \{ \tilde{\Phi}(1, 1, \tilde{\lambda}, r) - \Phi(1, 1, \lambda, r) \} \quad (14)$$

The limit  $\omega \rightarrow 0$  of the expression for  $\tau^{(2)}(\omega)$  can be shown to approach the value corresponding to the static dipolar polarizability which is 9/2.

### III. ELASTIC SCATTERING OF PHOTONS

The differential scattering cross section for low energy elastic scattering of photons with frequency  $\omega$ , by atoms is given by the dispersion formula [25, 26, 27]

$$\frac{d\sigma}{d\Omega} = a_0^2 (\epsilon \cdot \epsilon')^2 |M(\omega)|^2 = a_0^2 (\epsilon \cdot \epsilon')^2 |1 - P(\omega) - P(-\omega)|^2 \quad (15)$$

where  $a_0$  is the Bohr radius,  $M$  is the Kramers-Heisenberg matrix element,  $\epsilon$  and  $\epsilon'$  respectively are the initial and final polarization of photons and  $P(\omega)$  in atomic units is given as [27]

$$P(\omega) = -\frac{2}{3} \sum_n \frac{\langle g | \mathbf{p} | n \rangle \cdot \langle n | \mathbf{p} | g \rangle}{E_g - E_n + \omega}. \quad (16)$$

Here  $\mathbf{p}$  is the momentum operator and the summation is over the complete set of states including continuum states. It is useful to note from equation (15) that the differential cross section for coherent scattering of photons is just the Thompson cross section modified by the dynamic polarizability. In a similar fashion we can consider Raman scattering where initial and final states are different. The analytical expressions for  $P(\omega)$  were derived earlier using Schwartz and Teinman method [26] and Coulomb Greens function (CGF) formalism [27].

Using a slight variation of the formalism in the previous section we can easily calculate  $P(\omega)$ . In this case the infinite summation over the intermediate state in equation (16) is performed by defining a set of operators  $\mathbf{U}$  and  $\tilde{\mathbf{U}}$  such that,

$$\mathbf{p}|g\rangle = (\mathbf{U}H_0 - H_0\mathbf{U} + \omega\mathbf{U})|g\rangle \quad (17)$$

$$\mathbf{p}|g\rangle = (\tilde{\mathbf{U}}H_0 - H_0\tilde{\mathbf{U}} - \omega\tilde{\mathbf{U}})|g\rangle \quad (18)$$

and the expression for  $M$  will become

$$M(\omega) = 1 + \frac{2}{3} \left( \langle g | \mathbf{p} \cdot \mathbf{U} | g \rangle + \langle g | \mathbf{p} \cdot \tilde{\mathbf{U}} | g \rangle \right) \quad (19)$$

Now the evaluation of the infinite summation is reduced to the evaluation of the operators  $\mathbf{U}$  and  $\tilde{\mathbf{U}}$ . To obtain a similar expression as in the previous section, instead of going to momentum space representation [26, 27], we use the coordinate space representation of  $\mathbf{U}$  and  $\tilde{\mathbf{U}}$ . This is done by taking  $\mathbf{U} = \mathbf{r}u(r)$  and  $\tilde{\mathbf{U}} = \mathbf{r}\tilde{u}(r)$  and the equations (17) and (18) become

$$r \frac{d^2}{dr^2} u(r) + (4 - 2r) \frac{d}{dr} u(r) + (2\omega r - 2)u(r) = 2i \quad (20)$$

$$r \frac{d^2}{dr^2} \tilde{u}(r) + (4 - 2r) \frac{d}{dr} \tilde{u}(r) - (2\omega r + 2)\tilde{u}(r) = 2i \quad (21)$$

These differential equations has the same form which appeared in the previous section and the solutions can be written as

$$u(r) = -\frac{i}{2\omega^2} \Phi(1, 1, \lambda, r) \quad (22)$$

and

$$\tilde{u}(r) = \frac{i}{2\omega^2} \tilde{\Phi}(1, 1, \tilde{\lambda}, r) \quad (23)$$

Using this in the definition of  $U$  and  $\tilde{U}$  and substituting it in equation (19) we get a closed form expression for the Kramers-Heisenberg matrix element. It now takes the form

$$M(\omega) = 1 - \frac{4}{3\omega^2} \int_0^\infty dr e^{-2r} r^3 \left\{ \Phi(1, 1, \lambda, r) - \tilde{\Phi}(1, 1, \tilde{\lambda}, r) \right\} \quad (24)$$

If the incident photons are unpolarized and the polarization of the scattered photons are not observed the differential scattering cross section will take the standard form

$$d\sigma = r_0^2 \frac{1}{2} (1 + \cos^2 \theta) |M(\omega)|^2 d\Omega \quad (25)$$

#### IV. DISCUSSION AND CONCLUSION

The radial integrals in equations (14) and (24) can be done exactly [37]. For the numerical evaluation of various

integrals it is very convenient to define the following

$$\begin{aligned} I(p, q, \lambda, n) &= \int_0^\infty dr e^{-2r} r^n \Phi(p, q, \lambda, r) \\ &= n! \left( \frac{1-\lambda}{1+\lambda} \right)^{\frac{1}{\lambda}} \int_\lambda^1 ds \frac{(s+\lambda)^{p+\frac{1}{\lambda}} (s-\lambda)^{q-\frac{1}{\lambda}}}{(1+s)^{n+1}} \end{aligned} \quad (26)$$

and

$$\begin{aligned} \tilde{I}(p, q, \lambda, n) &= \int_0^\infty dr e^{-2r} r^n \tilde{\Phi}(p, q, \lambda, r) \\ &= n! \left( \frac{\lambda-1}{\lambda+1} \right)^{\frac{1}{\lambda}} \int_1^\lambda ds \frac{(\lambda+s)^{p+\frac{1}{\lambda}} (\lambda-s)^{q-\frac{1}{\lambda}}}{(1+s)^{n+1}}. \end{aligned} \quad (27)$$

With these, the Kramers-Heisenberg matrix element and the dipole dynamic polarizability respectively will take the following forms

$$M(\omega) = 1 - \frac{4}{3\omega^2} \left\{ I(1, 1, \lambda, 3) - \tilde{I}(1, 1, \tilde{\lambda}, 3) \right\} \quad (28)$$

$$\tau^{(2)}(\omega) = \frac{2}{3\omega^3} \left\{ \tilde{I}(1, 1, \tilde{\lambda}, 4) - I(1, 1, \lambda, 4) \right\} \quad (29)$$

This is the final form of the analytic expression for the matrix elements. We did not attempt to rewrite these in terms of well known special functions because the remaining integrals in equations (26) and (27) can be done very efficiently by a simple numerical integration.

The integrals in equation (10) is defined only for  $\text{Re}(q - 1/\lambda) > -1$ . But in order to calculate the matrix elements in equations (28) and (29) for all values of  $\omega$ , we require an analytical continuation of the definition of  $\Phi(\cdot)$  given in equation (10). It can be achieved by performing a partial integration in the definition given in equation (10) and this gives

$$\begin{aligned} \Phi(p, q, \lambda, r) &= \frac{1}{q+1-\frac{1}{\lambda}} [K(p, q+1, \lambda) + r\Phi(p, q+1, \lambda) \\ &\quad + \left( p + \frac{1}{\lambda} \right) \Phi(p-1, q+1, \lambda, r)]. \end{aligned} \quad (30)$$

This is a very important recurrence relation because (30) can be used to analytically continue the definition of these function for  $\text{Re}(q - 1/\lambda) > -2$ . We can also repeat this procedure to analytically continue the definition to other values of  $\text{Re}(q - 1/\lambda)$ . And for the numerical evaluation of various integrals as a function of radiation frequency  $\omega$  we have extensively used this relation. It is also very important to note the appearance of simple poles in the definition of  $\Phi(\cdot)$  when  $q = -1 + 1/\lambda$ . Since  $\lambda = \sqrt{1 - 2\omega}$ , this is due to the presence of intermediate resonances (when  $2\omega = 1 - 1/n^2$ , where  $n = 2, 3, \dots$ ) in

the expression for second order matrix element in the perturbation theory. This also leads to a strong frequency dependence of ac stark effect and scattering cross section.

Similarly for above one photon ionization threshold i.e.  $\omega > 1/2$ , a simple analytic continuation makes  $\lambda$  to be a purely imaginary number. Thus by this simple method, with the same analytic expression for the matrix element, we can numerically evaluate it for the whole physical range of photon frequency  $\omega$ . Displayed in table I are  $\tau^{(2)}(\omega)$  as a function of photon frequency in atomic units and are compared with values obtained by other methods. It is purely real for  $\omega < 1/2$ . For  $\omega > 1/2$  one photon ionization is possible and this makes the intermediate virtual state to lie in the continuum and  $\tau^{(2)}(\omega)$  becomes complex.

The dipole dynamic polarizability for photon energy below and above one photon ionization threshold is given in table I. Our values are compared with values obtained by Arnous et al. [17] using Coulomb Greens function method, but with an additional factor of  $\omega^{-2}$  in their result. It is very easy to see that this factor is missing from their result by taking the  $\omega \rightarrow 0$  limit. In this limit our result approaches the dc polarizability, which is

9/2. The values given in [17] also approaches to the same limit provided their results are multiplied by an overall factor of  $\omega^{-2}$ . Using the analytic continuation described in equation (30) we can calculate  $\tau^{(2)}(\omega)$  for  $\omega$  very close to intermediate resonance also. Our result also agrees with the second order level shift and width reported by Pan et al. [16]. From the values given in tables I and II it is useful to note the change in sign of the level shift when the photon frequency cross these resonance values. The values of Kramers-Heisenberg matrix element are given in tables II and III. They are in good agreement with the results of Gavrila [27].

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TABLE I: Comparison of the values of dipole dynamical polarizability in atomic units for both below and above one photon ionization threshold  $\omega = 0.5$ . In the limit  $\omega \rightarrow 0$ ,  $\tau^{(2)}(\omega)$  approaches the dc polarizability, which is -4.5.

$\omega$ (a.u)			$\omega$ (a.u)	Re $\tau^{(2)}(\omega)$		Im $\tau^{(2)}(\omega)$	
	This work	ABM <sup>a</sup>		This work	ABM <sup>a</sup>	This work	ABM <sup>a</sup>
0.001	4.50003	—	0.6	3.297	—	2.505	—
0.002	4.50011	—	0.7	2.493	—	1.408	—
0.02	4.51066	-4.51	0.8	1.915	1.915	0.850	0.8506
0.04	4.5429	-4.5431	1.0	1.205	1.205	0.362	0.3627
0.08	4.6775	-4.6776	2.0	0.275	0.275	0.023	0.0239
0.10	4.7843	-4.7843	3.0	0.117	—	0.004	—
0.20	5.9416	-5.9416	4.0	0.064	—	0.001	—
0.43	0.2971	—	5.0	0.041	0.041	0.0005	0.00057
0.46	3.9273	—	6.0	0.028	—	0.00027	—
0.465	3.0867	—	9.0	0.012	—	0.000049	—
0.477	1.2644	—	10	0.010081	0.01008	0.0000319	0.00003
0.478	1.9330	—					
0.489	0.6465	—					

<sup>a</sup>Values obtained using Coulomb Green function [17] by Arnous et al. with an additional  $\omega^{-2}$  factor as explained in section IV.

TABLE II: Kramers-Heisenberg matrix element  $M$  in atomic units for  $\omega$  below one photon ionization threshold. Comparison is made with values in Ref. [27].

$\omega$ (a.u)	This		$\omega$ (a.u)	This		$\omega$ (a.u)	This	
	Work	Ref. [27]		Work	Ref. [27]		Work	Ref. [27]
0.002	0.000018	—	0.376	77.8416	—			
0.02	0.0018	0.0018	0.38	15.3829	15.3828	0.481	4.0681	
0.04	0.0072	0.0072	0.4	2.6916	2.6916	0.484	0.6692	
0.06	0.0165	0.0165	0.429	0.0611	—	0.485	0.4490	
0.08	0.0299	0.0299	0.43	0.0549	0.0549	0.486	16.087	
0.10	0.0478	0.0478	0.44	3.1503	3.1503			
0.12	0.0708	0.0708	0.444	38.8927	—	0.488	1.2367	
0.14	0.0999	0.0999				0.489	0.1546	
0.16	0.1361	0.1361	0.445	32.2604	32.2603	0.49	6.6498	
0.18	0.1812	0.1812	0.453	2.3124	2.3124	0.491	1.2466	
0.20	0.2376	0.2376	0.464	0.2004	—	0.492	3.0681	
0.22	0.3091	0.3091	0.465	0.6674	0.6674	0.493	1.2572	
0.24	0.4016	0.4016	0.468	8.1693	8.1693	0.494	3.9947	
0.26	0.5246	0.5246				0.496	3.0200	
0.30	0.9507	0.9507	0.469	27.9814	27.9814	0.497	3.1600	
0.32	1.3752	1.3752	0.473	1.97857	1.9785	0.497	3.1600	
0.36	5.3036	5.3036	0.477	0.2876	—	0.498	0.7238	
0.37	15.763	15.763	0.478	0.4416	0.4416			

TABLE III: Real and Imaginary part of Kramers-Heisenberg matrix element for photons of energies above one photon ionization threshold ( $\omega > 0.5$ ) and comparison is made with values in Ref. [27].

$\omega$ (a.u)	Re $M$		Im $M$	
	This work	Ref. [27]	This work	Ref. [27]
0.6	1.1872	1.1872	0.9018	0.9018
0.7	1.22161	1.2216	0.6900	0.6900
0.8	1.22612	1.2261	0.5444	0.5444
0.9	1.21842	1.2184	0.4400	0.4400
1.0	1.20598	1.2059	0.3627	0.3627
2.0	1.10007	1.10007	0.0958	0.0958
3.0	1.05696	1.0569	0.0421	0.0421
4.0	1.03685	1.0368	0.0231	0.0231
5.0	1.02589	1.0258	0.0144	0.0144
6.0	1.01924	1.0192	0.00977	0.00977
7.0	1.01489	1.0148	0.00699	0.00699
8.0	1.01188	1.0118	0.00522	0.00522
9.0	1.00971	1.0097	0.00403	0.00403
10	1.0081	1.0081	0.00319	0.00319
20	1.00236	1.0023	0.00066	0.00066
30	1.00112	—	0.000262	—
40	1.00066	—	0.000133	—
50	1.00042	—	0.000075	—
90	1.00014	—	0.0000196	—